

# Hydrodynamic Interactions in Two Dimensions

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We measure hydrodynamic interactions between colloidal particles confined in a thin sheet of fluid. The reduced dimensionality, compared to a bulk fluid, increases dramatically the range of couplings. Using optical tweezers we force a two body system along the eigenmodes of the mobility tensor and find that eigen-mobilities change logarithmically with particle separation. At a hundred radii distance, the mobilities for correlated and anti-correlated motions differ by a factor of two, whereas in bulk fluids, they would be practically indistinguishable. We derive the two dimensional counterpart of the Oseen hydrodynamic tensor which quantitatively reproduces the observed behavior. These results highlight the importance of dimensionality for transport and interactions in colloidal systems and proteins in biological membranes.

When governing a world of lower dimensionality, the laws of physics give rise to intriguing phenomena. The reduced number of spatial dimensions usually results in stronger and longer ranged correlations. One example of this is long range Coulomb correlations and electron-lattice interactions which give rise to peculiar electronic and structural phase transitions in system of low dimensionality [1]. In a similar way, fluid flow propagators, mediating hydrodynamic interactions between suspended bodies, have logarithmic tails in two dimensions [2], giving rise to strong dynamical correlations [3].

When two particles suspended in a viscous fluid approach each other, propagation of fluid flow results in hydrodynamic interactions. As a consequence, a particle's motion resulting from a given stimuli, strongly depend on the entire instantaneous spatial configuration. Hydrodynamic interactions tend to favour correlated motions where every particle can move within the "slip-stream" of its neighbours [4]. This has consequences on many collective phenomena such as colloidal aggregation and gel formation where attractive interactions push to modify interparticle distances [5]. Moreover particles are always subject to stochastic thermal forces whose effects may be strongly influenced by hydrodynamic interactions, as happens in polymer dynamics [6], or for protein conformational changes [7, 8]. In three dimensional bulk fluids, the strength of hydrodynamic coupling decays as the inverse interparticle distance. This long range character makes hydrodynamic interactions quite effective in determining dynamical behavior and poses a number of numerical and theoretical challenges to the physical modeling of such phenomena.

The situation is even more dramatic when fluid flow is restricted to two dimensions, such as in cell membranes or soap films, and flow fields decay logarithmically with distance. It is important to note that spatially confin-

ing the particles to two dimensions is not sufficient to observe these long range tails. It is essential that the momentum flow is similarly restricted on a two dimensional plane. To this aim it is crucial that the bounding fluid has a much smaller viscosity than the film itself, a solid boundary would lead instead to hydrodynamic interactions decaying faster than in the 3D case [10].

The problem of two dimensional, single particle mobilities has received a lot of attention due to its relevance for Brownian motion in cellular membranes [9]. However, much less is known about the equally non trivial and important role of many body hydrodynamic interactions in two dimensional systems. Using video microscopy, Cheung and coworkers [3] observed long ranged spatial correlations between spheres floating in a free standing liquid film. Manybody effects were found to be significant even at very small concentrations, practically precluding the possibility of isolating hydrodynamic effects from indirect bulk measurements.

In this Letter we demonstrate how optical micromanipulation allows the direct observation of hydrodynamic interactions between an isolated pair of colloidal particles suspended in a free standing liquid film. Optical tweezers allow the positioning of two beads at varying separations, isolated from boundaries and other particles. The tweezers can also be used to simultaneously drive particles along any direction and directly probe the full mobility tensor. As a result, we observe a dramatic increase in the interaction range of hydrodynamic interactions due to the reduced dimensionality. On the other hand, we derive a two dimensional expression for the Oseen hydrodynamic tensor. Once the relevant far field boundary conditions are recognized, we can quantitatively describe experimental observations.

We are interested in determining particle motions under the action of an external force field. To the lowest

order in hydrodynamic interaction (large enough inter-particle distances), it is sufficient to know single particle quantities and in particular the mobility  $b$  and the far field Green's tensor  $\mathbf{G}(\mathbf{r})$  describing flow propagation:

$$\dot{\mathbf{R}}_i = b\mathbf{F}_i \quad (1)$$

$$\mathbf{u}(\mathbf{r}) = \mathbf{G}(\mathbf{r} - \mathbf{R}_i) \cdot \mathbf{F}_i \quad (2)$$

where  $\mathbf{u}(\mathbf{r})$  is the flow field produced by a force  $\mathbf{F}_i$  applied to a particle located in  $\mathbf{R}_i$ .

In a many body system the velocity of the  $i$ -th particle will be (within the superposition approximation) the sum of two contributions: the speed that it would have in the absence of other particles plus an “ambient” velocity obtained as the sum of all the fluid velocities independently produced at  $\mathbf{R}_i$  by other particles located at  $\mathbf{R}_j$ .

$$\dot{\mathbf{R}}_i = b\mathbf{F}_i + \sum_{j \neq i} \mathbf{G}(\mathbf{R}_i - \mathbf{R}_j) \cdot \mathbf{F}_j \quad (3)$$

In bulk three dimensional fluids,  $b$  is the inverse Stokes drag ( $1/6\pi\mu a$ ) while  $\mathbf{G}$  is the Oseen tensor [6] which, in cartesian coordinates, reads:

$$G_{3D}^{\alpha\beta}(\mathbf{r}) = \frac{1}{8\pi\mu r} \left[ \delta_{\alpha\beta} + \frac{r^\alpha r^\beta}{r^2} \right] \quad (4)$$

If we neglect stresses produced by the fluid bounding the film (air in our case) then the variations of flow properties across the film are negligible and dynamics is governed by a two dimensional Stokes equation. In other words, we only allow for in plane momentum flow and neglect contributions from the film surfaces. Working with relatively slow flows at the micron scale, the Reynolds number will be negligibly small allowing to express the momentum balance equation in the form of a two dimensional Stokes equation:

$$\mu \nabla^2 \mathbf{u}(\mathbf{r}) - \nabla p(\mathbf{r}) = -\mathbf{F}\delta(\mathbf{r})/h \quad (5)$$

When complemented by the incompressibility condition  $\nabla \cdot \mathbf{u} = 0$ , equation (5) can be rewritten as a biharmonic equation and solved for the propagator:

$$G_{2D}^{\alpha\beta}(\mathbf{r}) = \frac{1}{4\pi\mu h} \left[ \delta_{\alpha\beta} \left( \log \frac{L}{r} - 1 \right) + \frac{r^\alpha r^\beta}{r^2} \right] \quad (6)$$

where we have neglected  $1/r^2$  terms, whose coefficients depend on particle size and shape and boundary conditions on its surface. Such terms soon become negligible when moving away from the particle and play no role for hydrodynamic interactions between sufficiently separated particles. However, these terms determine the actual value of velocity on the particle boundary and therefore the single particle mobility  $b$ . The logarithmic term

appearing in (6) precludes the possibility of determining the integration constant  $L$  by imposing a vanishing velocity at infinity. The divergence of flow field signals the presence of a length scale beyond which some of the assumed approximations fails.

There are three approximations involved in the derivation of Eq. (5): 1) infinite size of the film, 2) negligible inertia, 3) negligible viscous drag on the interfaces. For each of them there is a length scale beyond which the solution in (6) is not self consistent with the assumed approximation. For the first approximation, the length scale is clearly the actual size of the film  $L_1$ . Following [9] we can impose a frictionless boundary on a ring of radius  $L_1$  and obtain the expression (6) for the propagator where  $L = L_1$ . For a stick boundary condition one gets  $L = L_1/\sqrt{e}$  [11]. In the second case, the flow field propagated by (6) fails to satisfy the negligible inertia approximation when the distance  $r$  is of order  $L_2 = \mu/\rho U$ , where  $\rho$  is the fluid density and  $U$  is the typical particle speed. However, inertial terms in the Stokes equation can be approximately taken into account by the Oseen method [12] and obtain a short distance ( $r \ll L_2$ ) expression for the propagator which is given by (6) with  $L = 4 \exp[1 - \gamma] L_2 \simeq 6.1 L_2$  ( $\gamma$  is the Euler-Mascheroni constant). At last, one can obtain from Saffman solution [9] that the momentum flow through film interfaces cannot be neglected for distances of the order of  $L_3 = h\mu/\mu'$  where  $h$  is the film thickness and  $\mu, \mu'$  are respectively the viscosities of the film and that of the bounding fluid. However, even in that case, expression (6) remains valid in a neighborhood  $r \ll L_3$  when we replace  $L$  with  $\exp[1/2 - \gamma] L_3 \simeq 0.9 L_3$ . When dealing with mesoscopic systems however, due to the very low Reynolds number involved,  $L_2$  is usually very large, a few hundred meters in our case.  $L_3$  is also quite big when the bounding fluid has a much lower viscosity than the film fluid, as in the present case where  $L_3 \sim 0.1\text{m}$ . Therefore, we expect that hydrodynamic interactions in free standing liquid films are usually dominated by the finite size of the membrane. The following discussion will remain valid even in other situations, provided one uses the relevant length scale  $L$ .

We can arrange all  $\mathbf{R}_i$  in a single  $2N$  dimensional vector and introduce the  $2N \times 2N$  hydrodynamic mobility tensor  $\mathbf{H}$  so that (3) reads:

$$\dot{\mathbf{R}} = \mathbf{H}(\mathbf{R}) \cdot \mathbf{F} \quad (7)$$

$$H_{ij}^{\alpha\beta}(\mathbf{R}) = \delta_{ij} \delta_{\alpha\beta} b + (1 - \delta_{ij}) G^{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) \quad (8)$$

We now have a two dimensional expression for the propagator (6) that can be used in (8) to get the many body mobility tensor as a function of the two parameters  $b$  and  $L$ . We will check the validity of our expression for  $\mathbf{H}$  by observing the dynamical behavior of two colloidal particles confined in a free-standing liquid film. In

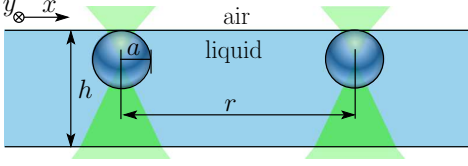


FIG. 1: (Color online) Trapping geometry. Two beads of radius  $a$  are optically trapped at a distance  $r$  in a liquid film of thickness  $h$ .

a two body system, if we choose the  $x$  axis along the joining line,  $x$  and  $y$  dynamics are naturally decoupled for symmetry reasons. We can then decompose  $\mathbf{H}$  into two  $2 \times 2$  mobility tensors operating on the subspaces of  $x$  and  $y$  coordinates. Any  $2 \times 2$  symmetrical matrix is diagonal in the coordinate system of the two eigenvectors  $(1, 1)$  and  $(1, -1)$ , corresponding respectively to a rigid translation and a stretching motion. Without losing any generality we can then assume that our mobility tensor is fully characterized by its four eigenvalues. Using expression (6) in (8) we can diagonalize the mobility tensor in the two body case and obtain the four eigenvalues as a function of interparticle distance  $r$ :

$$\lambda_{x\pm} = b \left[ 1 \pm \frac{1}{4\pi\mu hb} \log \frac{L}{r} \right] \quad (9)$$

$$\lambda_{y\pm} = b \left[ 1 \pm \frac{1}{4\pi\mu hb} \left( \log \frac{L}{r} - 1 \right) \right] \quad (10)$$

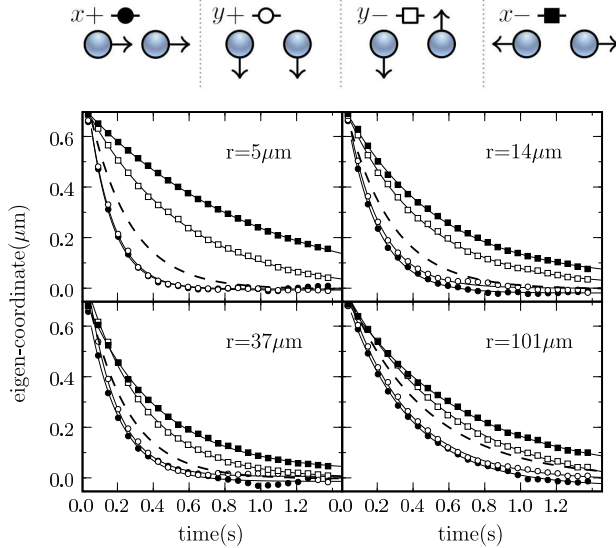


FIG. 2: Relaxation of eigen-coordinates. For each of the selected distances, we report the time evolution of the four eigen-coordinates relaxing to equilibrium after the applied perturbation. Solid lines are exponential fits. Dashed line is the average, single particle dynamics.

As in the three dimensional case, hydrodynamic inter-

actions produce a splitting in the spectrum of mobilities. The splitting is symmetric about the average single particle mobility and is larger for parallel than for perpendicular forcing. In three dimensions, the entity of the splitting decays as the inverse separation falling below 10% when particles distance grows beyond ten radii. The dependence on distance, in the two dimensional case, occurs only through a logarithmic term, which makes hydrodynamic interactions practically unavoidable.

In order to directly observe 2D hydrodynamic interactions, latex beads ( $2 \mu\text{m}$  diameter) are dispersed in a water-glycerol mixture with 0.2% wt SDS surfactant added. A thin film is obtained by sweeping the solution on a square frame (6 mm side) of nylon wires ( $60 \mu\text{m}$  thickness) [13]. Glycerol increases viscosity and slows down both drainage and evaporation, resulting in longer lived films. Starting with a 50% wt water/glycerol film and then heating to evaporate most of the water, we can obtain a very viscous film with a few micron thickness. We measured the film thickness  $h$  at the beginning and at the end of the reported experiment to be  $3.9 \mu\text{m}$ . Particles are imaged by a 40x NA 0.75 objective of an inverted optical microscope (Nikon TE2000-U). The same objective is used to focus the laser beam ( $\lambda=532 \text{ nm}$ ) diffracted off a spatial light modulator (SLM, Holoeye LCR-2500) into two, dynamically reconfigurable, optical traps [14, 15]. Axial confinement is by capillary force on the top surface of the liquid film (Fig. 1).

In contrast to previous studies of hydrodynamic interactions in 3D [4, 16, 17], where eigen-mobilities were extracted from the correlated fluctuations of optically trapped particles, we choose to measure the eigen-mobilities by directly exciting the four eigenmodes. For each of the four eigenmodes, we calculate two holograms producing two sets of traps slightly displaced along the eigencoordinate. The two holograms are alternatively displayed onto the SLM. The SLM is fully refreshed with the new hologram on a time scale (50 ms) which is much faster than the time scale of particle dynamics. The two particles will be then displaced from the new equilibrium positions, along the selected eigencoordinate. In our experiment, external forces are of optical and stochastic origin:

$$\mathbf{F}_i = -k\delta\mathbf{R}_i + \mathbf{S}_i \quad (11)$$

with  $k$  the trap strength and  $\delta\mathbf{R}_i = \mathbf{R}_i - \mathbf{R}_i^0$  the  $i$ th particle displacement from trap center  $\mathbf{R}_i^0$ . Assuming small displacements compared to interparticle distances and averaging over Brownian motion, we get from (7, 11):

$$\langle \delta\dot{\mathbf{R}} \rangle = -k\mathbf{H}(\mathbf{R}^0) \cdot \langle \delta\mathbf{R} \rangle \quad (12)$$

If the initial configuration corresponds to a small displacement along the  $n$ th eigenmode  $\langle \delta\mathbf{R}(0) \rangle = \epsilon\mathbf{e}_n$ , then (12) has the solution:

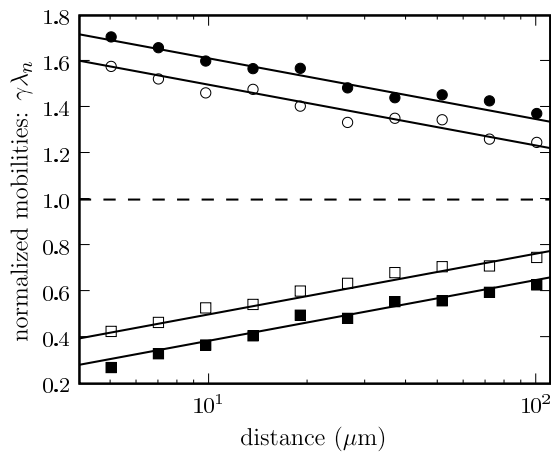


FIG. 3: Eigen-mobilities. The four eigen-mobilities of a two particle system arranged at different particles separations. To correct for variations in trapping power, for each distance mobilities have been normalized to their average.

$$\langle \delta \mathbf{R} \rangle = \epsilon \exp[-k\lambda_n t] \mathbf{e}_n \quad (13)$$

and we can directly obtain the corresponding mobility  $\lambda_n$  by monitoring the amplitude of the  $\mathbf{e}_n$  mode relaxing to equilibrium. We choose to normalize the eigenvectors such that the corresponding eigencoordinates give the center of mass position, for the rigid modes, and the half distance (along  $x$  or  $y$ ) for stretching modes.

We choose ten logarithmically spaced interparticle separations between  $5 \mu\text{m}$  and  $100 \mu\text{m}$  and drive the two particles back and forth (eight times) along each of the four eigenmodes. Particle coordinates were digitally extracted from video frames at 144 Hz. Eigencoordinates were then computed and averaged over the eight iterations. Fig. 2 shows the time evolution of the four eigen-coordinates at four selected interparticle distances. The two  $x$  and  $y$  rigid motions are much more mobile than the corresponding stretching motions. This behavior remains clearly visible up to the highest investigated distance (100 particle radii). The four eigen-mobilities corresponding to the four probed modes can be extracted by fitting the eigen-coordinate dynamics to the exponential law in (13). To correct for variations in trap strength  $k$ , for each distance, we normalize the four obtained decay rates to their average value  $kb$ . The relative eigen mobilities are shown in Fig. 3 as a function of the particle separation. The strength of hydrodynamic coupling, reflected in the splitting of mobilities, decays logarithmically slow with distance. Still at a separation of 100 radii, particles move twice as fast when forced along the same direction rather than in the opposite. At the same large separation, three dimensional mobilities would only differ by 1%.

The four data sets in Fig. 3 can be very well fitted by (9) leaving  $L$  and the adimensional mobility  $b^* = 4\pi\mu hb$

as the only free parameters. We obtain as best fit parameters  $b^* = 8.7$  and  $L = 2.1 \text{ mm}$ , the corresponding fitting curves are shown as straight lines. As expected, the relevant length scale  $L$  is determined by the film finite size. In particular a sticky boundary condition on a ring inscribed in the film frame ( $L_1 = 3 \text{ mm}$ ) would give an  $L = 1.8 \text{ mm}$ . For the same boundary condition, the single particle mobility  $b$  can be calculated for a cylinder of height  $h$ , the film thickness [11]. Using our particle radius  $a$  as the cylinder radius we obtain  $b^* = \log(L_1/a) - 1 = 7.0$  which compares reasonably well with the corresponding fitted value. We do not expect the two values to be in better agreement since the mobility  $b$  depends on the details of the boundary conditions on the particle surface. We anticipate that, when moving to multiparticle systems, the mobility of long wavelength eigenmodes will diverge linearly with the number of particles, rather than logarithmically as in the 3D case. As a first consequence of that, we could transport, at the same speed, any number of beads, using the same total amount of laser power. Moreover, the crossover to underdamped propagating modes on a linear chain of trapped particles predicted in [17], could be experimentally verified much more easily.

We have directly measured hydrodynamic interactions between colloidal particles in a sheet of viscous fluid. The reduced dimensionality, compared to the bulk 3D fluids, results in stronger and longer ranged hydrodynamic couplings which are quantitatively reproduced by a two dimensional version of Oseen hydrodynamic tensor. The observed interactions constitute a general model for diffusion and interactions of proteins in biological membranes.

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